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THE EFFECT OF SOLID OXIDANT ADDITIVIES
ON DIFFUSION BURNING OF POLYMERS IN AIR

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Foreign Technology Division
Wright-Patterson Air Force Base, Ohio

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Block	Italic	Transliteration	Block	Italic	Transliteration
А а	<i>А а</i>	A, a	Р р	<i>Р р</i>	R, r
Б б	<i>Б б</i>	B, b	С с	<i>С с</i>	S, s
В в	<i>В в</i>	V, v	Т т	<i>Т т</i>	T, t
Г г	<i>Г г</i>	G, g	У у	<i>У у</i>	U, u
Д д	<i>Д д</i>	D, d	Ф ф	<i>Ф ф</i>	F, f
Е е	<i>Е е</i>	Ye, ye; E, e*	Х х	<i>Х х</i>	Kh, kh
Ж ж	<i>Ж ж</i>	Zh, zh	Ц ц	<i>Ц ц</i>	Ts, ts
З з	<i>З з</i>	Z, z	Ч ч	<i>Ч ч</i>	Ch, ch
И и	<i>И и</i>	I, i	Ш ш	<i>Ш ш</i>	Sh, sh
Й й	<i>Й й</i>	Y, y	Щ щ	<i>Щ щ</i>	Shch, shch
К к	<i>К к</i>	K, k	Ъ ъ	<i>Ъ ъ</i>	"
Л л	<i>Л л</i>	L, l	Ы ы	<i>Ы ы</i>	Y, y
М м	<i>М м</i>	M, m	Ь ь	<i>Ь ь</i>	'
Н н	<i>Н н</i>	N, n	Э э	<i>Э э</i>	E, e
О о	<i>О о</i>	O, o	Ю ю	<i>Ю ю</i>	Yu, yu
П п	<i>П п</i>	P, p	Я я	<i>Я я</i>	Ya, ya

* ye initially, after vowels, and after ъ, ь; e elsewhere.
 When written as Ѣ in Russian, transliterate as yě or Ѣ.
 The use of diacritical marks is preferred, but such marks
 may be omitted when expediency dictates.

THE EFFECT OF SOLID OXIDANT ADDITIVES ON DIFFUSION BURNING OF POLYMERS IN AIR

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(Moscow)

The rate of diffusion burning (w) of liquid fuels (in cylindrical pipes) or cores of solid fuels in a gaseous oxidizer increases considerably with a decrease in the core diameter and with an increase in oxygen concentration. The absolute value of w is very small; for example, for kerosene burning in air, $w = 0.15 + 0.08$ mm/s at 1 at and 6 + 8 mm tube diameter [1]. Very little study has been done on the pressure dependence.

In contrast, the burning rate (u) of mixtures of solid fuels with solid oxidizers depends only slightly on the diameter (if it is not too small) and composition of the ambient gas, but usually is substantially dependent on pressure. The absolute value of u is at least one order higher than that of w for solid fuels [2].

For a number of problems it is of interest to observe a transition from the first mode of burning to the second with the introduction into the fuel of gradually increasing additions of solid oxidizers. In this paper such a transition is observed during the addition of ammonium perchlorate (APC) to polymethyl methacrylate (PMMA).

PROCEDURE OF EXPERIMENTATION

Powdered PMMA ($\sim 3 \mu$) and APC were mixed on tracing paper with a rubber plug for one hour. Cylindrical charges 6-12 mm in diameter and 7-15 mm in height were pressed from the obtained mixture. The relative density of the charges was 0.95-0.99.

Tests were carried out in air at atmospheric pressure on an installation which allowed us to measure the mass burning rate (m_{cm} , g/cm²·s) with the aid of an elastic element and strain gauges [3]. A flexible steel plate (elastic element) with strain gauges attached to it was rigidly fastened at one end, while the studied sample was placed on the other end. The variation in weight of the sample changes the deflections of the plate and the resistance of the strain gauges, which is recorded by a strain recorder and an oscillograph. In order to obtain a flat burning surface the sample was blown with a stream of air parallel to the lateral surface of the sample. Ignition was accomplished by a Nichrome spiral which did not touch the specimen.

TEST RESULTS

Periodic two-stage burning was observed for compositions with a large excess of fuel. Due to the air's oxygen, the initial burning is that of pure fuel (PMMA). In this case the surface layer is enriched with an oxidizer, producing a mixture which, under these conditions is capable of burning as a result of its own oxidizer. This mixture ignites at a certain moment, followed by the combustion of the organic fuel, then another ignition, etc. It should be noted that each composition has its own characteristic time (τ) between ignitions, depending on the dispersion of the APC and which diminishes as the weight portion of the oxidizer (χ_{ox}) in the mixture increases (Table 1).

With a certain sufficiently large oxidizer content (χ_{ox}^*) the mixtures begin to burn in one state due to their own oxidizer

Table 1.

Z_{OK}	τ , s for APC of the following fractions:	
	1270 μ	9 μ
0.1	19.2	8.0
0.2	8.4	7.2
0.3	4.5	6.5
0.4	2.0	5.0

(i.e., burning occurs under the burning conditions of concentrated mixtures).

These mixtures correspond to the upper concentration limits of burning of condensed mixtures (critical mixture).

For coarsely dispersed APC the value of χ_{OH}^* is ~ 0.6 , while for finely-dispersed APC it is ~ 0.75 . In this case the mass burning rates are 0.035 and 0.068 g/cm²·s, respectively.

It is necessary to note that a region of unstable burning, where it is extinguished after one-two cycles, lies between the regions of two-stage burning and burning under the conditions of condensed mixtures. For coarse APC this region lies in the range of $0.4 < \chi_{OH} < 0.6$, while for fine APC $0.5 < \chi_{OH} < 0.75$.

Let us now examine in detail the effect of χ_{OH} on the value of the mass burning rate in the region of two-stage burning. The values of the mass rates are presented in Table 2.

Table 2.

Z_{OK}	m_{CH} , g/cm ² ·s for APC of the following fractions:	
	1270 μ	9 μ
0	0.00175 —	0.00175 —
0.1	0.0020/0.00202 (1)	0.0020/0.00201 (1)
0.2	0.0026/0.0025 (1)	0.0025/0.0024 (1)
0.3	0.0033/0.0033 (1)	0.0036/0.0029 (1)
0.4	0.0046/0.0045 (1)	0.0040/0.0036 (1)
0.5	—/—	0.0060/0.0050 (1)

Note: The lines 0.0020/0.00202 (1) - consist of the experiment data - 0.0020 - and values calculated by the corresponding formula - 0.00202 (1).

It is seen from Table 2 that the burning rate increases monotonically with an increase in the APC fraction; moreover, the numerical values of the rates are higher on the coarse oxidizer than on the fine.

DISCUSSION OF THE RESULTS

It is possible to propose a simple theoretical model of two-stage burning. We will assume that for such mixtures, the first to burn out is the pure fuel (PMMA) at a rate m_r . After a certain time there is ignition of the critical mixture, i.e., the mixture corresponding to the upper concentration limit of burning at a rate m^* .

Let us assume that the weight fraction of the oxidizer in the original mixture is equal to x_{OH} , while the weight fraction of the fuel $x_r = 1 - x_{OH}$. The portion of the fuel (x_r'), which burns due

to APC is determined from the relationship $\frac{y_{OK}}{x_r} = \frac{y_{OK}^*}{1 - y_{OK}^*}$. Accordingly,

a portion of the PMMA which burns due to the air's oxygen, is equal to $1 - \frac{y_{OK}}{y_{OK}^*}$. The characteristic burnout time for the PMMA is

equal to $\frac{1 - \frac{y_{OK}}{y_{OK}^*}}{m_r}$, while the ignition time is correspondingly equal to $\frac{\frac{y_{OK}}{y_{OK}^*}}{m^*}$.

Based on this the expression for the mass burning rate of the mixture will have the following form:

$$m_{cm} = \frac{1}{\frac{1 - \frac{y_{OK}}{y_{OK}^*}}{m_r} + \frac{y_{OK}}{y_{OK}^*} \frac{1}{m^*}} \quad (1)$$

where m_r and m^* are taken from the experiment.

The mass rate of the mixture, calculated by formula (1), is in good agreement with experiment (see Table 2).

In this model it is assumed that the oxidizer is burnt in the critical mixture composition together with a corresponding amount of fuel. The increase in the combustion rate of PMMA, with the introduction of the oxidizer additive, is due to the fact that the burning rate of the critical mixture is one order greater than that of diffusion burning of pure PMMA.

However, the burning rate of PMMA can also be increased if we introduce additives which, even though they do not react with the fuel, are gasified at a rate which considerably exceeds that of the diffusion burning of PMMA. Table 3 shows data on the effect of the easily gasified ammonium bicarbonate and ammonium chloride additives on the burning rate of PMMA (the data for APC are also given for comparison).

Table 3.

Percent of addi- tives	m_{cm} for the various additives		
	NH_4Cl 200—300 μ	NH_4HCO_3 200—300 μ	NH_4ClO_4 9 μ
0	0,00175	0,0175	0,00175
3	0,00206	0,00205	0,00203
5	0,0022	0,00209	0,0021
7	0,021	—	—
10	0,00175	0,0023	0,0022
15	Does not burn	0,0024	Does not burn
20	—	0,0023	0,0025
30	—	He горит	0,0036

In essence, both these additives are inert diluents and they decrease the heat of combustion only for the PMMA.¹ In addition, the gasification products of these additives (HCl, CO₂, and H₂O) dilute the reacting mixture (this problem is analogous to that concerning the drop in burning rate when the percent of oxygen in the ambient air is decreased). With small percents of additives NH₄Cl and NH₄HCO₃ the increase in the gasification rate plays a decisive role, while dilution has only a slight effect. Thus, with an NH₄Cl content up to ~5% and NH₄HCO₃ up to ~10% these additives increase the burning rate of PMMA to the same degree as NH₄ClO₄ (Table 3). However, with a further increase in the percent of NH₄Cl or NH₄HCO₃ the effect of dilution becomes the determining factor and the burning rate begins to drop right down to extinction.

In addition to the burning rate our attention is drawn to the frequency of ignitions observed during burning. Let us examine the case where a coarse oxidizer was added, where we can assume that each crystal of the oxidizer (together with the attached layer of fuel) ignites individually.

To obtain the expression for the time interval τ between ignitions, we will examine the burning of the sample with diameter D. The number of oxidizer particles per unit length of the sample is equal to $\frac{3}{2} \cdot \frac{D^3 \cdot \rho_{ox}}{d^3 \rho_{cm}}$ (here d - diameter of the oxidizer particle; ρ_{cm} and ρ_{ox} - density of the mixture and oxidizer). When a sample burns at an average mass rate m , the time between ignitions will be

$$\tau = \frac{2}{3} \cdot \frac{d^3}{D^2} \cdot \frac{\rho_{ox}}{\rho_{cm}} \cdot \frac{1}{m_{cm}} \quad (2)$$

¹The rated heat of NH₄Cl combustion due to the air's oxygen is one orders lower than that of hydrocarbons. We were unable to ignite in air either NH₄Cl or NH₄HCO₃.

In the case of the addition of fine PKhA the tests have shown that the ignition engulf the entire end of the sample and that the mass of mixture, consumed during the ignition, decreased¹ with a decrease in χ_{OK} . Thus, it is possible to examine a simple model where it is assumed that the time between ignitions is determined by the burning rate of a characteristic layer of mixture having the thickness $\frac{a}{u^*}$ (where a - thermal conductivity, u^* - linear combustion rate of critical mixture). The excess of PMMA, which is equal to

$$\rho_{CM} \cdot \frac{a}{u^*} \left(1 - \frac{\chi_{OK}}{\chi_{OK}^*} \right),$$

is burned out first, followed by the amount of critical mixture, which is equal to

$$\rho_{CM} \cdot \frac{a}{u^*} \cdot \frac{\chi_{OK}}{\chi_{OK}^*}.$$

The time between ignitions is equal to

$$\tau = \rho_{CM} \cdot \frac{a}{u^*} \left(\frac{1 - \frac{\chi_{OK}}{\chi_{OK}^*}}{m_r} + \frac{\frac{\chi_{OK}}{\chi_{OK}^*}}{m^*} \right). \quad (3)$$

The values of τ calculated by formula (2) for a coarse oxidizer (with $D = 8$ mm) and by formula (3) for a fine oxidizer ($a \approx 0.5 \times 10^{-3}$ cm²/s), are in good agreement with experiment (Table 4).

¹This can be evaluated indirectly from a recording of the plate vibration amplitude on an oscillogram at the moment of ignition. When $\chi = 0.5$ this amplitude is 21 mm, on the average, and when $\chi = 0.1$ it is only 3.5 mm.

Table 4.

Oxidizer %	τ , s for APC of the following fractions:	
	1270 μ	9 μ
10	19.2/21.8 (2)	8.0 7.53 (3)
20	8.4 8.35 (2)	7.2 6.66 (3)
30	4.5 3.7 (2)	6.5 5.75 (3)
40	2.0 2.2 (2)	5.0 4.73 (3)
50	—	3.7 3.62 (3)

Note: The lines 19.2/21.8 (2) - consist of the experiment data - 19.2 - and the values calculated by the corresponding formula - 21.8 (2).

CONCLUSIONS

1. It is shown that when APC is added to PMMA the rate of combustion of PMMA in air is increased considerably.
2. Mixtures of PMMA with 40-50% APC burn in air in two stages. The portion of pure PMMA is burned out first, followed by the ignition of the portion of the PMMA-APC mixture whose composition corresponds to the upper concentration limit of burning in an inert atmosphere, after which the cycle is repeated many times. In the intermediate region (between the region of two-stage burning and burning due to its own oxidizer) the burning is intermittent: the mixtures are extinguished after one-two cycles.
3. Simple theoretical models of two-stage burning are proposed which permit one to calculate the burning rates and frequency of ignitions for similar compositions.

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